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Effect of charge density wave (CDW) on magnetization in manganite systems

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Abstract : The manganite system exhibits colossal magneto-resistance which is presumed to be explained by the lattice distortion in addition to the double exchange mechanism. In the present communication we consider a simple Hamiltonian consisting of the hopping term arising due to the itinerant electrons of Mn atom in presence of Charge-Density-Wave (CDW) distortion and the external magnetic field in the e_g band. The nested pieces of the Fermi surface with strong electron-phonon interaction brings about the formation of a CDW with an accompanying periodic lattice distortion. The induced ferromagnetism in the e_g band originates due to the more localized t_{2g} electrons. The appropriate electron Green's functions are calculated leading to the calculation of the magnetization and CDW gap. The magnetization and CDW gap are solved numerically and self-consistently. The results are discussed taking into account different model parameters of the system.

Keywords : Electron-phonon interaction, magnetically ordered materials, exchange and super exchange.

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1. Introduction

The manganese oxides with perovskite structures $Re_{1-x}A_xMnO_3$ (Re = La, Pr, Nd and A = Sr, Ca) have attracted high scientific interest due to colossal magneto-resistance (CMR) [1,2]. The prototypes $La_{1-x}(Sr, Ca)_xMnO_3$ are since long the protagonists of the 'double exchange' mechanism [3]. Replacing in $La^{3+}Mn^{3+}O_3$, a tirivalent La^{3+} ion by a divalent earth alkali ion (Sr^{2+}, Ca^{2+}) requires an additional electron from the manganese for the binding. The result is a homogeneous valence mixture of the manganese ion $\left(Mn^{3+}_{1-x}, Mn^{4+}_{x}\right)$. The three $3d$–t_{2g} electrons of Mn^{4+} are considered as more or less localized forming a local S = 3/2 spin. The fourth electron in Mn^{3+} is of $3d$–e_g type and is itinerant. It is assumed that it interacts *via* intra-shell Hund's rule coupling ('double exchange mode') [4–6] with the S = 3/2 spins. $LaMnO_3$ undergoes the cooperative Jahn-Teller (J-T) transition below 780 K of the orthorhombic structure and lifts the degenerated e_g orbital, which orders such that nearest neighbour z-axis of $d_{3z^2-r^2}$ is alternately aligned with 90° or of antiferro-type order in the C-plane. The type-1 antiferromagnetic structure in $LaMnO_3$ has been interpreted by the strong ferromagnetic super exchange interaction acting in the C-plane with weak antiferromagnetic interaction perpendicular to the plane [7]. In this respect, if $d_{3z^2-r^2}$ orbital of the e_g band is stabilized, oxygen ions at apex, perpendicular to the basal plane approach closer to the central cation, hence the crystal undergoes to the tetragonal symmetry.

The e_g electrons interact with each other and with distortions of the surrounding lattices *i.e.* with phonons. The later interaction has been argued to be unusually strong in the CMR materials, essentially because the e_g electrons on a given Mn atom are strongly coupled to distortions of the surrounding O_6 octahedron [8–10]. In the pseudo cubic materials such as $La_{1-x}Sr_xMnO_3$, the strong coupling is due, in part, to the J-T effect. A local distortion, in which some Mn-O bonds get shorter and

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others get longer, breaks the local cubic symmetry and therefore splits the degeneracy of the e_g levels on that site. If only one electron is present, then it will, in the absence of hybridization, reside in the lower level and gain energy. The J-T effect leads to a large frozenin lattice distortion in $LaMnO_3$ [11,12] and has been argued to be important in the doped materials as well [9,10]. In the doped material a breathing mode distortion in which all six Mn-O bond lengths change by the same amount, has also been argued to be important [9,13]. The applicability of these considerations to the layered manganites is unclear, because in them the cubic symmetry has already been broken by the crystal structure. The electron-lattice coupling in the layered materials requires further attention. Recently, we have studied the interplay of ferromagnetism and static band J-T distortion present in e_g levels alone in manganite system [14]. In another communication, we have considered a model for the manganite system in which there exists a static J-T distortion in the e_g band and Heisenberg type interaction in t_{2g} core band in presence of a weak hybridization between the itinerant e_g electrons and the more localized t_{2g} core electrons [15]. The unusual properties of the CMR effect are regarded as a cooperative phenomena associated with the structural change due to the atomic displacements competing with the magnetic interactions and the charge fluctuations between different valencies of the manganese cations. Though J-T interaction is believed to play crucial role to explain CMR effect, we attempt here to study the manganite systems in presence of charge density Wave (CDW) type periodic lattice distortion in stead of static Jahn-Teller distortion. In Section 2 we discuss a model Hamiltonian for the manganite system. The Section 3 deals with the calculation of the Green's functions and the expressions for magnetization and CDW gap. Finally we discuss the results in Section 4 and conclusions in Section 5.

2. Formalism

We consider here the itinerant e_g electrons of Mn atoms in presence of the induced ferromagnetic order in the same e_g electrons, although the strong Hund's rule coupling aligns the localized spins in the t_{2g} state. In the simplified model under consideration, we assume the existence of a direct Heisenberg type exchange interaction between the e_g electrons to be responsible for induced ferromagnetic state. Furthermore, calculations are performed in the mean field approximation accounting for the magnetic exchange interaction in the Ising limit. Since the e_g band is doubly degenerate a charge density wave (CDW) distortion is invoked to remove the degeneracy. An external magnetic field drastically reduces the magnetic state and drives the system from insulating ferromagnetic state to a metallic ferromagnetic state. The aim of the calculation is to study the interplay between the transitions from the paramagnetic to the ferromagnetic state and the structural transition induced by the periodic lattice distortion. The different terms in the model Hamiltonian are detailed below.

We consider a model system in which the e_g electrons of the Mn atoms interact with the lattice as well as between themselves.

$$H_c = \sum_{k,\sigma} (\in (k) - \mu - B\sigma) c^{\dagger}_{k\sigma} c_{k\sigma} , \tag{1}$$

where H_c represents the non-interacting electronic energy in a two-fold degenerate (e_g) band with single particle energy $\in(k)$ and chemical potential μ. The operators $c^{\dagger}_{k\sigma}(c_{k\sigma})$ being the creation (annihilation) operator of the electron of spin σ and energy $\in(k)$. Here B is the external magnetic field acting in the spin direction σ where $\sigma = +1$ for spin up and $\sigma = -1$ for spin down.

The Fermi surface instability leads to the nested piece of Fermi surface giving rise to the charge density wave (CDW) state by opening up a gap ($2\Delta_c$) in the lattice driving the metallic system either to a semiconductor or to an insulator. The low dimensionality of the system provides nested pieces of the Fermi surface which gives rise to the electron-hole symmetry in the energy spectrum. $\in_{k+Q} = -\in_k$ where Q is the CDW nesting wave vector and $\in_k$ is the electronic energy. The nesting property with strong electron phonon interaction brings about the formation of CDW with an accompanying periodic lattice distortion (PLD). Fermi surface nesting may be present in higher dimensional systems, but may not be perfect. The electron-phonon interaction in a metallic system can be reduced to an effective electron-electron interaction and that can be attractive only under some appropriate condition with strength V being proportional to the square of the electron-phonon interaction strength such that

$$H_{e-e} = -V \sum_{k,k',\sigma,\sigma'} c^{\dagger}_{k+Q\sigma} c^{\dagger}_{k'-Q\sigma'} c_{k'\sigma'} c_{k\sigma} . \tag{2}$$

In order to describe the ground state of the system as the CDW insulator, the Q'th Fourier component of the charge density or equivalently the phonon amplitude can be taken as order parameter $\Delta_c = -V < \rho_a >$ where $\rho_a = \sum_{k,\sigma} c^{\dagger}_{k+Q\sigma} c_{k\sigma}$. The CDW state has a super periodicity such that Q becomes the new reciprocal lattice vector with a corresponding reduced Brillouin zone (RBZ). Therefore, the mean field Hamiltonian for the CDW state can be written as

$$H_{\mathrm{CDW}} = \Delta_c \sum_{k,\sigma}^{\mathrm{RBZ}} \left(c^{\dagger}_{k+Q\uparrow} c_{k\uparrow} + c^{\dagger}_{k+Q\downarrow} c_{k\downarrow} \right), \tag{3}$$

where

$$\Delta_c = -VN(0) \sum \left[\left\langle c^{\dagger}_{k\uparrow} c_{k+Q\uparrow} \right\rangle + \left\langle c^{\dagger}_{k\downarrow} c_{k+Q\downarrow} \right\rangle \right]. \tag{4}$$

Here, Δ_c is the CDW gap parameters and $N(0)$ is the density of state of the conduction electrons at the Fermi level.

The Heisenberg exchange interaction between the spins of e_g electrons at the i-th and j-th atomic sites in the Ising limit can be writen as

$$H_M = -JM \sum \left[c^{\dagger}_{k\uparrow} c_{k\uparrow} - c^{\dagger}_{k\downarrow} c_{k\downarrow} \right], \tag{5}$$

where the ferromagnetic order parameter is $M = - <S^z>$ and J is the effective interaction energy. The electron hopping is present in a ferromagnetic metallic system. The exchange energy per atom is comparable to the kinetic energy and it attains the magnitude of ~1 eV by interacting with all other electrons in the system.

The Coulomb interaction exists only within the same e_g electronic orbitals and the strength of interaction is same for both the orbitals. The Coulomb interaction due to the intinerant e_g electron densities can be written as

$$H_1 = U \sum_{k,\sigma} \left(c^{\dagger}_{k\uparrow} c_{k\downarrow} c^{\dagger}_{k\downarrow} c_{k\uparrow} \right), \tag{6}$$

where the Coulomb interaction strength U is assumed to be non-zero and isotropic constant (independent of momentum). The e_g electrons move in an average ferromagnetic field which is calculated from H_I in Hartree-Fock mean field approximation and given below

$$H_I = U \sum \left(n_{\uparrow} c^{\dagger}_{k\downarrow} c_{k\downarrow} + n_{\downarrow} c^{\dagger}_{k\uparrow} c_{k\uparrow} \right), \tag{7}$$

where $n_{\uparrow}$ and $n_{\downarrow}$ are defined by

$$n_{\uparrow} = \sum \left\langle c^{\dagger}_{k\uparrow} c_{k\uparrow} \right\rangle, \quad n_{\downarrow} = \sum \left\langle c^{\dagger}_{k\downarrow} c_{k\downarrow} \right\rangle. \tag{8}$$

Hence, the manganite system in presenece of charge density wave (CDW) distortion ferromagnetism and the external magnetic field in the e_g band can be described by the total Hamiltonian H given by

$$H = H_c + H_{\mathrm{CDW}} + H_M + H_I. \tag{9}$$

3. Calculation of electron Green's functions

The double time single particle electron Green's functions are calculated by equations of motion method of Zubarev [16]. The elelctron Green's functions are defined as

$$A_1(k,\omega) = \left\langle\left\langle c_{k\uparrow}; c^{\dagger}_{k\uparrow} \right\rangle\right\rangle_{\omega},$$

$$A_2(k,\omega) = \left\langle\left\langle c_{k+Q\uparrow}; c^{\dagger}_{k\uparrow} \right\rangle\right\rangle_{\omega},$$

$$B_1(k,\omega) = \left\langle\left\langle c_{k\downarrow}; c^{\dagger}_{k\downarrow} \right\rangle\right\rangle_{\omega},$$

$$B_2(k,\omega) = \left\langle\left\langle c_{k+Q\downarrow}; c^{\dagger}_{k\downarrow} \right\rangle\right\rangle_{\omega}. \tag{10}$$

The Green's functions are calculated and written as

$$A_1(k,\omega) = \frac{\omega - \bar{\epsilon}_{1k\uparrow}}{2\pi |D_1(\omega)|} \qquad A_2(k,\omega) = \frac{}{2\pi |D_1(\omega)|}$$

$$B_1(k,\omega) = \frac{\omega - \bar{\epsilon}_{1k\downarrow}}{2\pi |D_2(\omega)|} \qquad B_2(k,\omega) = \frac{\Delta^c}{2\pi |D_2(\omega)|} \tag{11}$$

where

$$D_1(\omega) = (\omega - \epsilon_{1k\uparrow})(\omega - \bar{\epsilon}_{1k\uparrow}) - \Delta_c^2,$$

$$D_2(\omega) = (\omega - \epsilon_{1k\downarrow})(\omega - \bar{\epsilon}_{1k\downarrow}) - \Delta_c^2, \tag{12}$$

and

$$\epsilon_{1k\uparrow} = \epsilon(k) - \mu - B + \frac{1}{2} JM + U < n_{\downarrow} >,$$

$$\bar{\epsilon}_{1k\uparrow} = -\epsilon(k) - \mu - B + \frac{1}{2} JM + U < n_{\downarrow} >,$$

$$\epsilon_{1k\downarrow} = -\epsilon(k) - \mu + B - \frac{1}{2}JM + U < n_\uparrow >,$$

$$\bar{\epsilon}_{1k\downarrow} = -\epsilon(k) - \mu + B - \frac{1}{2}JM + U < n_\uparrow >. \quad (13)$$

The occupation numbers $n_{k\sigma}$ for the conduction electrons are found to be

$$n_{k\uparrow} = \frac{1}{2E_k}\sum_{i=1}^{2}\left[\left(E_k - (-1)^i(\epsilon(k) - \mu)\right)f(\omega_i)\right],$$

$$n_{k\downarrow} = \frac{1}{2E_k}\sum_{i=3}^{4}\left[\left(E_k - (-1)^i(\epsilon(k) - \mu)\right)f(\omega_i)\right], \quad (14)$$

where $E_k^2 = (\epsilon(k) - \mu)^2 + \Delta_c^2$,

$$\omega_1, \omega_2 = -B + \frac{1}{2}JM + U < n_\downarrow > \pm E_k$$

$$\omega_3, \omega_4 = -B - \frac{1}{2}JM + U < n_\uparrow > \pm E_k, \quad (15)$$

where the Fermi function in general is given by $f(y) = \frac{1}{e^y + 1}$. The reduced magnetization $m = M/N$ in the e_g band of the manganite system is given by

$$m = -\sum(n_{k\uparrow} - n_{k\downarrow}), \quad (16)$$

where the occupation numbers $n_{1\uparrow}$ and $n_{1\downarrow}$ are difined by eq. (8). The sum over the momenta of the electrons in the conduction band is replaced by the integration over energy of the electron $\epsilon_0(k)$ with integration limit from $-W/2$ to $+ W/2$ as $\sum_k = \int_{-W/2}^{+W/2} N(\epsilon_0)d\epsilon_0$. Here, $N(\epsilon_0)$ is the model density of states [17] and $2D$ is the total band width W of the conduction band. To simulate the strong energy dependence of $N(\epsilon_0)$ around the center of the band, we take

$$N(\epsilon_0) = N(0)\sqrt{1 - \left|\frac{\epsilon_0}{D}\right|}\ln D \quad (17)$$

where $N(0)$ is unperturbed density of state of the free electron and ϵ_0 is the kinetic energy of the conduction band.

The dimensionless CDW gap reduces to

$$= -g_1\sum_k \frac{1}{2E_k}[f(\omega_1) - f(\omega_2) + f(\omega_3) - f(\omega_4)]. \quad (18)$$

The physical quantities involved in the calculations are the external magnetic field (B), band energy ($\epsilon(k)$), chemical potential (μ), the strength of the electron-lattice interaction (g_1), the lattice distortion (e), the effective interaction energy (J), the temperature (T), coulomb interaction U and the conduction band width (W). These quantities are scaled with respect to the band energy (W). The dimensionless parameters are given by

$$y = \frac{\epsilon(k)}{W},\ z = \frac{\mu}{W},\ b = \frac{B}{W},\ t = \frac{k_B T}{W},\ e = \frac{\Delta_c}{W},$$

$$g_1 = N(0)V,\ g = \frac{J}{W},\ u = \frac{U}{W}. \quad (19)$$

4. Results and discussions

In the manganite system $La_{1-x}Sr_xMnO_3$ there occurs a static elastic strain in the electron density of the degenerate e_g electron band. Due to strong Hund's rule coupling the t_{2g} electrons align ferromagnetically and this magnetism in t_{2g} band induces a weak ferromagnetism in e_g band. The lattice distortion in the form of charge density wave (CDW) distortion is expected to suppress or enhance the ferromagnetism in e_g band. In the present communication, we want to study the effect of CDW distortion on the magnetization and ferromagnetic Curie temperature. The dimensionless parameters involved in the calculations are the magnetization (m), CDW gap (e), CDW coupling strength (g_1), magnetic coupling strength (g), chemical potential (z), temperature (t), magnetic field (b) and the Coulomb interaction (u).

(A) Induced ferromagnetism in e_g band :

The magnetization (m) in eq. (16) and CDW gap (e) in eq. (18) are solved individually and self-consistently for a set of coupling parameters $g_1 = 0.0989$, $g = 0.299$ in absence of external magnetic field and chemical potential and their temperature dependence is shown in Figure 1. The coupling constants are so chosen that the ferromagnetism (FM) transition temperature t_c and the CDW lattice distortion temperature t_d satisfy the condition $t_c > t_d$. Here $t_c = 0.025$ and $t_d = 0.023$. The magnetization m and CDW gap e exhibit mean-field behavior. When the two parameters interplay, both are suppressed

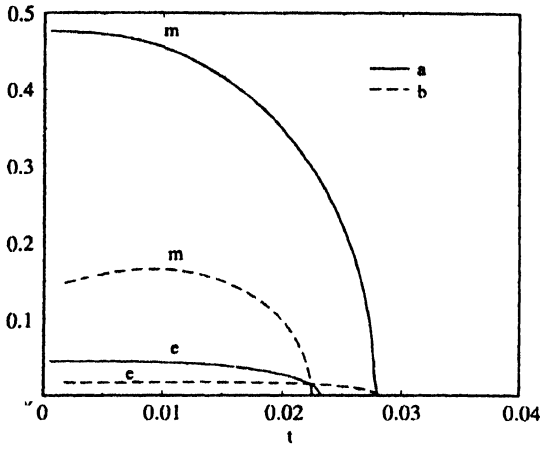

Figure 1. (a) Individual plots of m *vs* t and e *vs* t (for fixed values of $b = 0$, magnetic coupling $g = 0.299$ and CDW coupling $g_1 = 0.0989$ respectively, (b) self-consistent plots.

considerably in presence of each other as compared to their individual values (see solid line in Figure 1). The magnetization is suppressed from $m = 0.47$ to 0.15 and the CDW gap is suppressed from $e = 0.045$ to 0.02. Moreover, the distortion temperature t_d is enhanced, while the FM t_c is suppressed in the coexistence phase leading to the condition $t_d > t_c$. Thus, the CDW periodic distortion suppresses the FM metallic phase.

The effect of external magnetic field on the CDW gap e is shown in the Figure 2. The external field

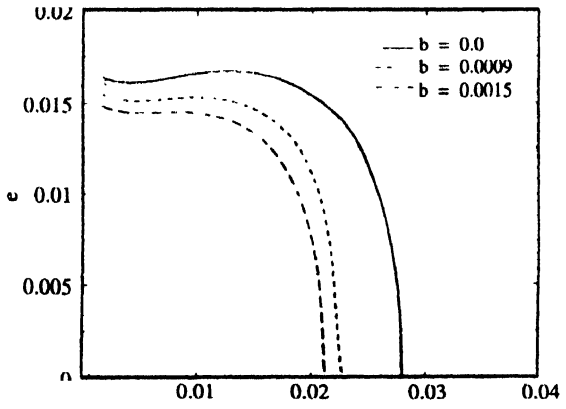

Figure 2. Self-consistent plots of e *vs* t for fixed values of $g = 0.299$, $g_1 = 0.0989$ and for different values of magnetic field $b = 0$, 0.0009, 0.0015.

suppresses the CDW gap throughout the temperature range and decreases the distortion temperature t_d. This suppression of the CDW gap changes the magnetization in the same range of temperature for the given external field as shown in Figure 3. When distortion temperature $t_d > t_c$, the magnetization is unaffected by external magnetic field *i.e.* for $b = 0$. When the external magnetic field suppresses the distortion temperature t_d which is closer to the FM transition temperature t_c, the magnetization shows new features. The magnetization increases with increase of external field up to the distortion

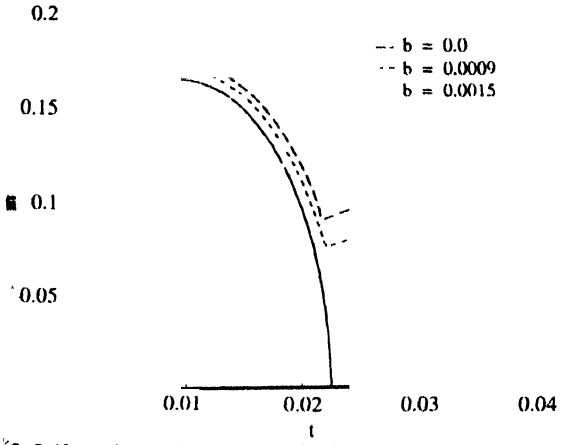

Figure 3. Self-consistent plots of m *vs* t for fixed values of $g = 0.2990.185$, $g_1 = 0.0989$ and for different values of magnetic field $b = 0$, 0.0009, 0.0015.

temperature and exhibits mean-field behavior. The transition temperature t_c is suppressed slightly on the application of the magnetic field in the coexistence phase. For temperature $t > t_c$, the magnetization increases with temperature in the paramagnetic phase.

(B) Paramagnetism in e_g band :

The motion of the e_g electrons on the background of the t_{2g} core electrons can be described by double exchange Hamiltonian. The e_g electron on site j is constrained by the strong Hound's coupling to be parallel to the core spin s_j on that site. The Physics may be expressed by using the exchange Hamiltonian

$$H'_M = -J_H \sum_{\alpha,j} s_{\alpha j}.S_{\alpha j} \tag{20}$$

where $S_{\alpha j}$ is the e_g electron spin at site j with α being band index ($\alpha = 1, 2$). Here $J_H > 0$ implies ferromagnetic coupling.

The eq. (20) implies a connection between magnetism in core t_{2g} band and the transport of e_g electron. In the Ising limit the Hamiltonian given in eq. (20) can be written as

$$H'_M = \frac{J_H M'}{2} \sum_{\alpha,k} \left(c^{\dagger}_{\alpha k\uparrow} c_{\alpha k\uparrow} - c^{\dagger}_{\alpha k\downarrow} c_{\alpha k\downarrow} \right), \tag{21}$$

where the mean field magnetization due to the t_{2g} band is given by the parameter $h = J_H M'$. The paramagnetic magnetization m^e in the e_g band is calculated from the expression given in eq. (21).

The magnetization (m^e) and the CDW lattice strain (e) are solved self-consistently and their temperature

dependences are shown in the Figure 4. The CDW coupling g_1 enhances both the CDW strain and the

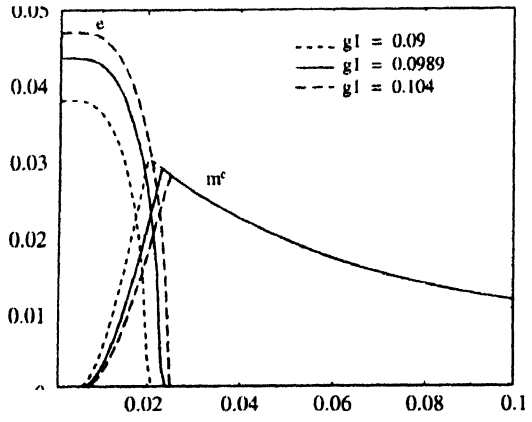

Figure 4. Self-consistent plots of m *vs* t and e *vs* t for fixed values of $b = 0$, $h = 0.008$, and different values of CDW coupling $g_1 = 0.09, 0.0989, 0.104$.

distortion temperature t_c separating the distorted and undistorted phases. For temperatures $t > t_d$, the paramagnetic magnetization shows usual temperature dependence and remains unaffected by the CDW strain. But for the temperatures $t < t_d$, the magnetization reduces drastically and attains zero magnetization at $t = 0$ inside the distorted phase of the system. Moreover, the CDW coupling decreases the magnetization in the distorted phase.

The effect of a mean-field magnetization (h) of the t_{2g} band on the CDW strain and the paramagnetism is shown in Figure 5. The mean-field h does not bring

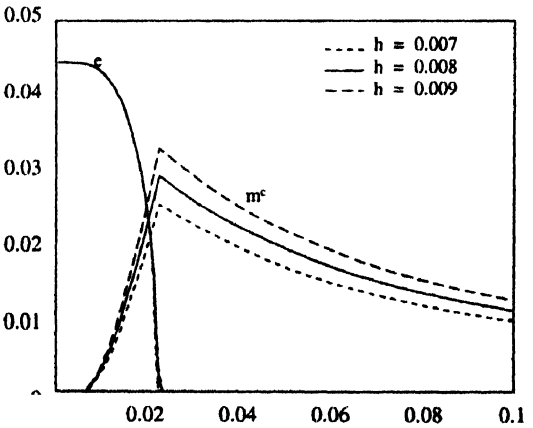

Figure 5. Self-consistent plots of m *vs* t and e *vs* t for fixed values of $b = 0$, $g_1 = 0.0989$ and different values of $h = 0.007, 0.008, 0.009$.

about any change in the CDW strain. However, the background mean-field h of the t_{2g} band enhances the paramagnetic magnetization m^c of the e_g band throughout the temperature range. The magnetization is considerably reduced for temperatures $t > t_d$.

5. Conclusions

We consider here a model study of the manganite system in which a periodic lattice distortion and induced ferromagnetism exists in the e_g band. When the two phases interplay with each other, both phases are suppressed considerably. The external magnetic field suppresses the CDW strain and decreases the distortion temperature t_d. This suppression of CDW strain changes the magnetization in the same range of temperatures for the given external field. For the distortion temperature t_d greater than t_c, the magnetization is unaffected by the external magnetic field. When the external field suppresses the distortion temperature t_d close to the FM transition temperature, the magnetization increases almost linearly for temperatures $t > t_c$. The motion of the e_g electrons on the background of the ferromagnetically ordered t_{2g} core electrons exhibits paramagnetism. The CDW coupling enhances the lattice strain in the e_g band. As a result, the paramagnetism is totally suppressed in the CDW distorted phase. The background ferromagnetic field h due to the t_{2g} band has no influence on the CDW lattice strain. The paramagnetism in the e_g band still remains suppressed in the distorted phase. However, the field h increases the magnetization in the paramagnetic phase for temperatures $t > t_d$. Thus, we conclude that the CDW distortion has a profound influence on the magnetization of the e_g band. These effects can be seen clearly by studying the temperature dependence of density of states of the e_g band and the magnetoresistivity.

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